Multi-ferroics with Polarization in Laminate Composites

Shuxiang Dong, Jie-Fang Li, and D. Viehland[*], Materials Science & Engineering,

Virginia Tech, Blacksburg, VA 24061

Abstract

It has been discovered that laminate composites of longitudinally magnetized magnetostrictive and transversely poled piezoelectric layers (a L-T laminate composite) have a giant magneto-electric (ME) effect under a low magnetic bias. The ME voltage coefficient is over 110 mV/Oe at a magnetic bias H=500 Oe. This value is 5-10 times higher than that previously reported for transverse magnetized/ transverse polarized (T-T) laminates of the same layer compositions at the same bias. In this paper, we also report the magneto-elasto-electric bi-effect equivalent circuit of the L-T laminate composite and the corresponding theoretical formula of the magneto-electric voltage coefficient.

Introduction

The magneto-electric ME effect is a polarization P response to an applied magnetic field H, or conversely a magnetization M response to an applied electric field E [1]. ME effects have been reported in single phase materials [2-4]; however to date, a ME material with significant coupling has yet to be found. ME behavior has also been reported in piezoelectric/ magnetostrictive composites [5-15]. Various composite types have been studied; including ceramic-ceramic particle and laminate ones. High magneto-electric effects have previously been found in transverse (thickness) magnetized/ transverse (thickness) polarized or T-T laminates, but only under high magnetic bias [8,11].

However, we have identified an inherent flaw in the T-T laminate ME composite. Gauss's law requires both the magnetic and electric fluxes to be continuous across the interface between piezoelectric and magnetostrictive layers, i.e., $\varepsilon_1 E_1 = \varepsilon_2 E_2$ and $\mu_1 H_1 = \mu_2 H_2$. Thus, in the composite configuration, the magnetic potential drops across the dielectric layer, and the electric potential across the magnetic one. This is the opposite of what is required for significant ME effects at low bias.

We have overcome this limitation by reconfiguration of the laminate composite as shown in Fig.1(a), such that the electric and magnetic potentials both drop across appropriate layers. This long type laminate consists of longitudinally magnetized Terfenol-D and transversely poled piezoelectric layers, relative the principle (longitudinal) vibration direction – *a (L-T) laminate composite*. This configuration is also compatible with the tensor property matrix for Terfenol-D, as its longitudinal strain is 8x times higher than the transverse. Thus, the L-T laminate in Fig.1(a) achieves: (i) drop of appropriate flux lines across respective layers, (ii) continuity of both flux lines, and (iii) maximum tensor property combinations.

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14. ABSTRACT

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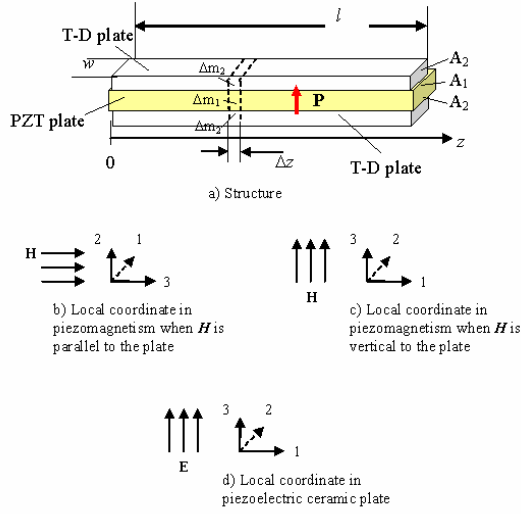| 16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	18. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
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Figure 1. (a) Design of our L-T piezomagnetic/piezoelectric laminated composite; (b) local coordinates in the magnetostrictive layer when H is applied parallel to plate; (c) local coordinates in magnetostrictive layer when H is vertical to the plate; and (c) local coordinates in piezoelectric layer.

Magneto-elasto-electric Bi-effect Coupling

Terfenol-D (TbDyFe$_2$) is a giant (positive) magnetostrictive material [17,18]. Although the strain S induced by H is magnetostrictive ($S = \lambda H^2$, where λ is the magnetostrictive coefficient), it has a dc-biased or pseudo-linear piezomagnetic coefficient [10, 17]. The maximum piezomagnetic coefficient occurs at the inflection point of the quadratic S-H response. Correspondingly, a linear piezomagnetic equation can be used to describe the magneto-elastic effect of Terfenol-D.

Following Figures 1a, L-T laminates were made of two Terfenol-D plates and a soft Pb(Zr$_{1-x}$Ti$_x$)O$_3$ (PZT-5) plate. The length and width of each layer was 12.7 mm and 6 mm, respectively. The total cross area A of the three layers laminated composite was 6x2.5 mm^2. The thickness of the PZT layer was from 0.5 to 1.0 mm. A d.c magnetic bias H$_{dc}$ was applied along the longitudinal direction for the L-T laminate (Fig.1b), and along the transverse direction for the T-T (Fig.1c). In the linear approximations, the longitudinal and transverse piezomagnetic (d$_{33,m}$ and d$_{31,m}$) coefficients of the piezomagnetic constitutive equations, and the transverse piezoelectric coefficient d$_{31,p}$ of the piezoelectric constitutive equations are mutually coupled through strain S(z) and stress T(z) in their thickness directions. Thus, application of H along the length direction of the magnetostrictive layer (see Fig.1b) puts the piezoelectric one into forced oscillation, generating a voltage across its thickness (see Fig.1d). Applying Newton's 2nd law of motion to the laminate and finding analogous electrical parameters [16,17], a magneto-elasto-electric bi-effect equivalent circuit can be found as shown in Fig. 2. The ME voltage coefficient of the L-T laminate derived from this equivalent circuit is

$$\left|\frac{dV}{dH}\right|_{L-T} = \frac{n(1-n)A d_{33,m} d_{31,p}^2}{\varepsilon_{33}^T s_{11}^E [n s_{11}^E (1-k_{31,p}^2) + (1-n) s_{33}^H]} \tag{1}$$

where s_{11}^E and s_{33}^H are the elastic compliances of the piezoelectric and magnetostrictive layers respectively, and $k_{31,p}$ and ε_{33}^T are electromechanical coupling coefficient and dielectric constant at constant stress of the piezoelectric material. Using the appropriate materials parameters in table I, we estimate the maximum

value of $\left| \dfrac{dV}{dH} \right|_{L-T}$ for the L-T laminate of Terfenol-D and PZT to be 54 mV/Oe at a thickness ratio of piezomagnetic plate to composite plate of n =0.64.
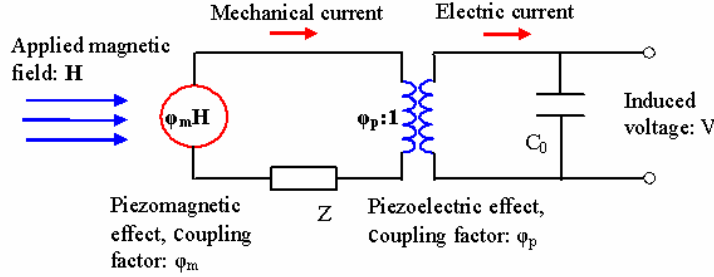

Figure 2. Magneto-elastic-electric bi-effect equivalent circuit of the L-T laminate, where $Z = -\dfrac{1}{2} j \overline{\rho}\, \overline{\upsilon}\, Actg\ \dfrac{kl}{2}$ is the impedance, $\varphi_m = A_2 \dfrac{d_{33,m}}{s_{33}^H}$ is the magnetoelastic coupling factor, $\varphi_p = \dfrac{w d_{31,p}}{s_{11}^E}$ is the piezoelectric coupling factor, $C_0 = \dfrac{lw}{t_1} \varepsilon_{33}^S$ is the piezoelectric capacitance. This equivalent circuit is important for understanding the magnetoelectric coupling of the laminate. An applied magnetic field **H,** which also acts as a "mechanical voltage", induces a "mechanical current" $\dot{u}$ via the magneto-elastic effect with a coupling factor φ_m. In turn, this results in an electrical voltage V across the piezoelectric layer due to electromechanical coupling. A transformer with a turn-ratio of φ_p can be used to represent the electromechanical coupling in the circuit. The *I* produced across the piezoelectric layer is then the electromechanical coupling current.

Table I. Electromechanical and magnetoelastic materials parameters for [1]Terfenol-D, PZT, and <001>-oriented PMN-PT crystals.

	[2]$d_{33,m}$ or $d_{33,p}$	[2]$d_{31,m}$ or $d_{31,p}$	s_{11}^H or s_{11}^E	s_{33}^H or s_{33}^E	k_{33}	k_{31}	ε_{33}^T	χ
Terfenol-D	1.2×10^{-8} Wb/N	-5.8×10^{-9} Wb/N	125×10^{-12} m^2/N	40×10^{-12} m^2/N	0.7			5
PZT 5 ceramic	600 pC/N	-310 pC/N	14.8×10^{-12} m^2/N	16.7×10^{-12} m^2/N	0.65	0.30	2300	1
<001> PMN-PT	2820 pC/N	-1330 pC/N	69.0×10^{-12} m^2/N	119×10^{-12} m^2/N	0.92	0.59	[3]2134	1

[1]From ETREMA Products, Inc., 2500 N. Loop Drive, Ames, Iowa 50010, U.S.A.
[2]Magnetostrictive constants, $d_{33,m}$ and $d_{31,m}$ are related to magnetic field bias applied to piezomagnetic material, Terfenol-D;
[3]Measured value after assembling.

Experimental Results

The induced voltage across the PZT plate was then measured for various H_{dc} (magnetic bias) and H_{ac} (magnetic exciting signal) at a working frequency of 10^3 Hz, using a lock-in amplifier method. Fig.3 shows the ME voltage coefficient as a function of H_{dc}, at a constant H_{ac}=1 Oe. For the L-T laminate, the ME voltage coefficient increases dramatically with increasing H_{dc}, reaching a maximum of ~57 mVp/Oe at H_{dc}=500 Oe. For the T-T laminate, in this bias range, the ME voltage coefficient is ~10 mV/Oe. Although, a maximum value of ~58 mVp/Oe was observed for $H_{dc} \geq 3000$ Oe, consistent with previous investigations of T-T laminates of similar layer compositions [8,11].
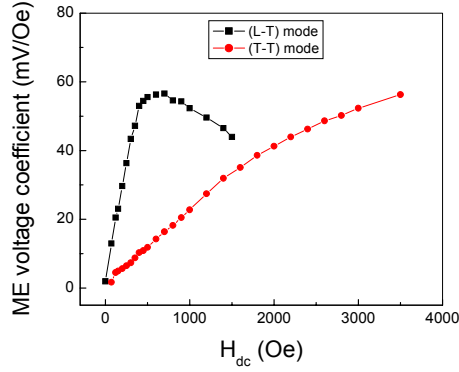

Figure 3. Magneto-electric voltage coefficient as a function of H_{dc}. Measurements were performed using a H_{ac}=1 Oe (amplitude) at a measurement frequency of 10^3 Hz.

The induced ME voltage was a linear function of H_{ac} for both the L-T and T-T laminates, as shown in Figure 4 with H_{dc}=500 Oe. The induced voltage was much higher for the L-T laminates. Using a H_{ac}=1.2 Oe, the ME voltage for the L-T laminate was nearly 70 mV. Whereas, for the T-T laminate, the corresponding ME voltage was only about 12 mV.
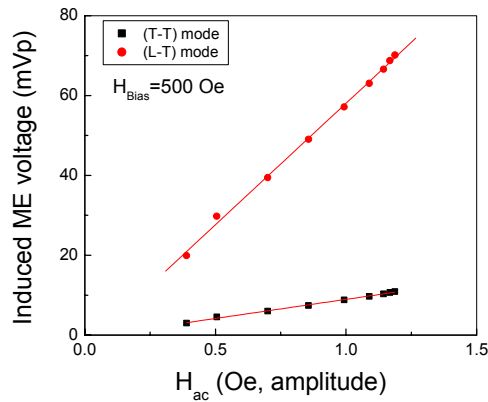

Figure 4. Induced magneto-electric voltage as a function of H_{ac} for Terfenol-D/PZT composite. The measurement frequency was 10^3 Hz.

The magnetoelectric coupling coefficient k_{me} is related to the ratio of the electric to magnetic energy densities. From equation (1), k_{me} can be given as

$$k_{me}^2 = \frac{\varepsilon E^2}{\mu H^2} = \frac{\varepsilon}{t_1^2 \mu} \left(\frac{n(1-n)Ad_{33,m}d_{31,p}^2}{\varepsilon_{33}^T s_{11}^E [ns_{11}^E(1-k_{31,p}^2) + (1-n)s_{33}^H]} \right)^2 \tag{2}$$

where t_1 is the thickness of piezoelectric plate. In the range $H_{dc}<1000$ Oe, this ratio is 100-400x higher for the L-T laminate relative to the T-T. From the square root of the ratio, the ME coupling at $H_{dc}=500$ Oe was determined to be 0.15 for the L-T laminate.

Even higher ME coupling was found for L-T laminates of Terfenol-D and <001>-oriented *0.7*Pb(Mg$_{1/3}$Nb$_{2/3}$)O$_3$-*0.3*PbTiO$_3$ (PMN-PT), as PMN-PT is known to have significantly higher piezoelectric constants and electromechanical coupling coefficients than PZT (see table I). Fig.5 shows the induced voltage as a function of H_{ac} for an L-T laminate of Terfenol-D and PMN-PT. Using a $H_{ac}=1$ Oe, the ME voltage was 11 mV, 32 mV and 110 mV for biases of 0, 70 and 500 Oe, respectively. The ME coupling at $H_{dc}=500$ Oe was determined to be 0.28. The values for k_{me} are close to the thermodynamic limits for the L-T laminate, which is given by $k_{me}^2 = k_{31,piezo}^2 \times k_{33,Terfenol-D}^2$.
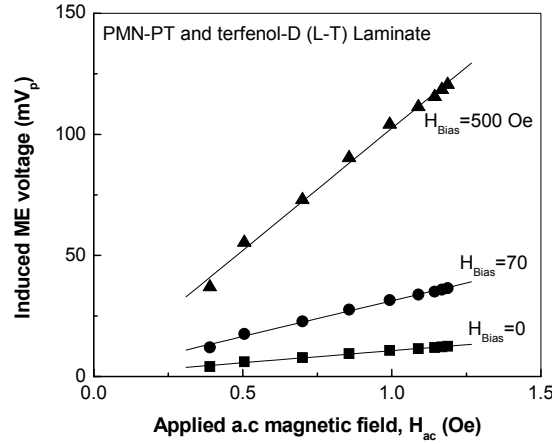

Figure 5. Induced magneto-electric voltage as a function of H_{ac} for the L-T laminate of Terfenol-D and <001>-oriented PMN-PT. Data are shown for various H_{dc}. The measurement frequency was 10^3 Hz.

Conclusions

In this paper, a L-T mode piezomagnetic/piezoelectric laminated composite has been designed using an approximate magneto-elasto-electric (or bi-effect) equivalent circuit, prototyped and characterized. The results for the L-T laminates demonstrate much promise for ME sensors. They offer good magnetic and electric energy coupling, excellent sensitivity, a signal that has a linear coupling between *E* and *H* that is independent of motional impedance, and in addition operates under zero or low dc bias. Also, the results show that the L-T mode has a ME voltage coefficient 5-10x higher than that of the conventional T-T mode.

Acknowledgements

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